Capture of Carbon Dioxide from Flue Gas Using a Cyclic Alkali Carbonate-Based Process

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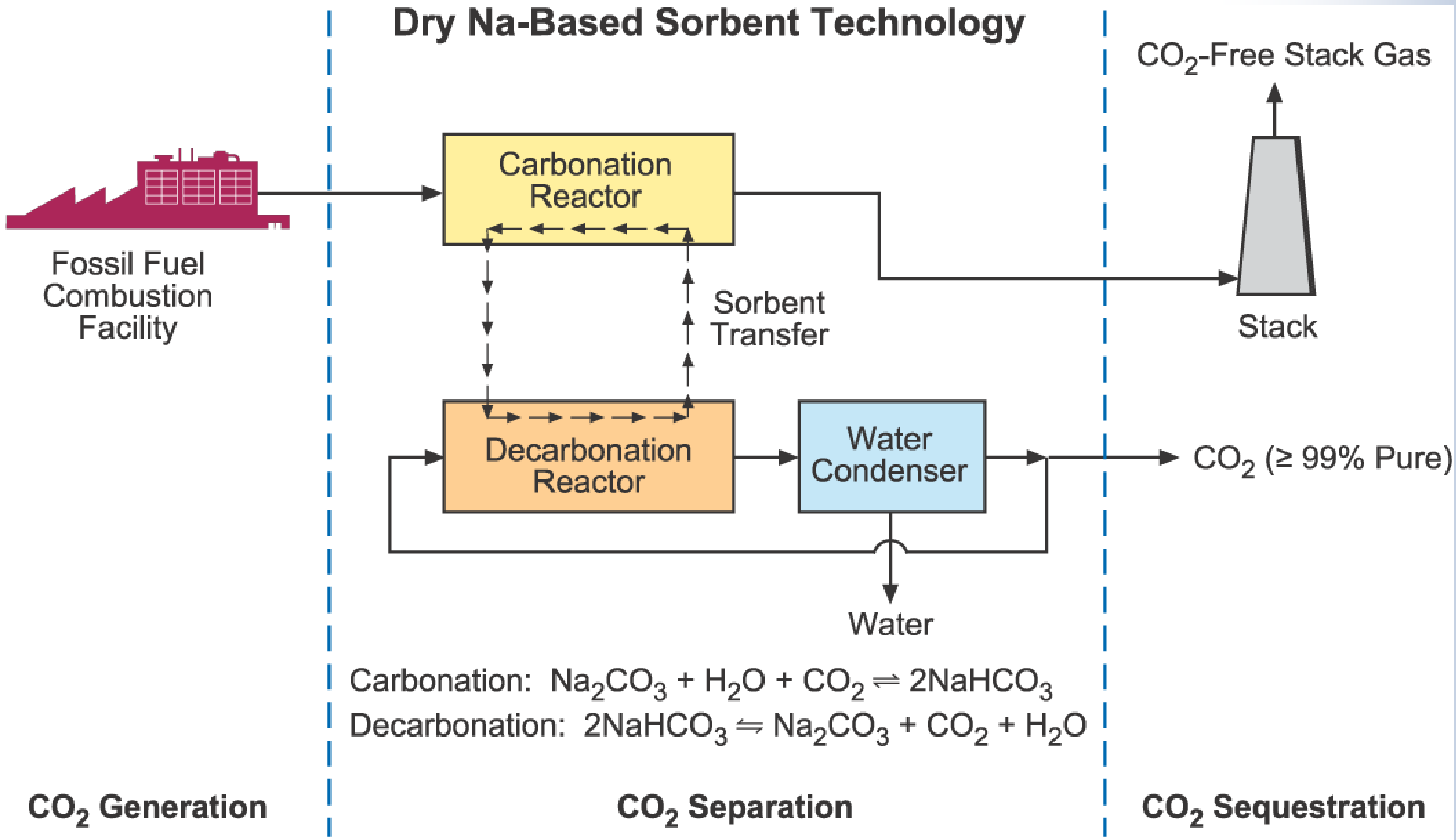
Objectives

To develop a carbon dioxide separation technology that is

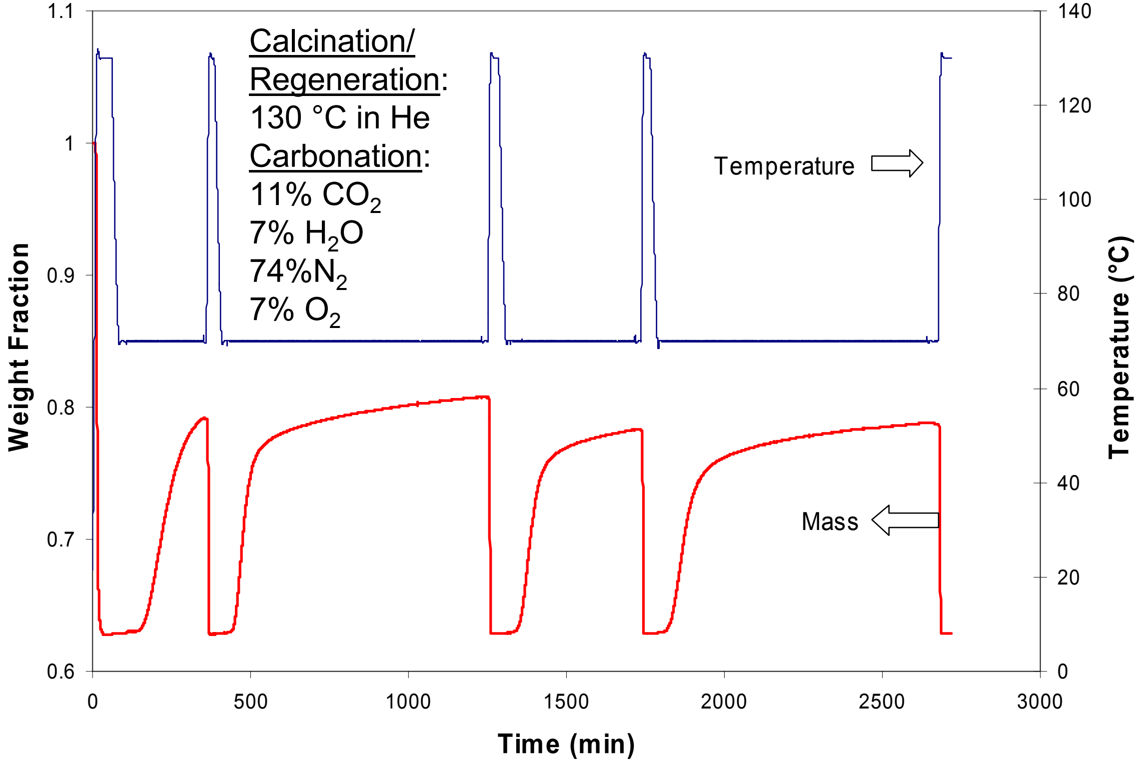
- Regenerable sorbent-based
- Applicable to both coal and natural gas-based power plants
- Applicable as a retrofit to existing plants, as well as to new power plants
- Compatible with the operating conditions in current power plant configurations
- Relatively simple to operate
- Less expensive than currently available technologies

Integration of the "Dry Carbonate" Process in a Combustion Facility



Concept Evaluation

(Sodium Bicarbonate Sorbent – “Baking Soda”)



- Inexpensive CO_2 getler identified
- Getler is readily regenerated
- Low temperature process
- Convenient for flue gas treatment

Materials Screened

- Sodium bicarbonate (SBC) – $NaHCO_3$
  - Grade 1
  - Grade 2
  - Grade 3
  - Grade 5
  - Spherical
- Trona--$Na_2CO_3 \cdot NaHCO_3 \cdot 2H_2O$
  - Grade T-50
  - Grade T-200
- Potassium Carbonate – K_2CO_3
  - Analytical Grade
  - Commercial Grade
  - Jet-milled
- Supported Sorbents
  - 40% K_2CO_3/60% support
  - 10% K_2CO_3/90% support
  - 20% Na_2CO_3/80% support
  - 40% Na_2CO_3/60% support

Sorbent Characterization and Testing

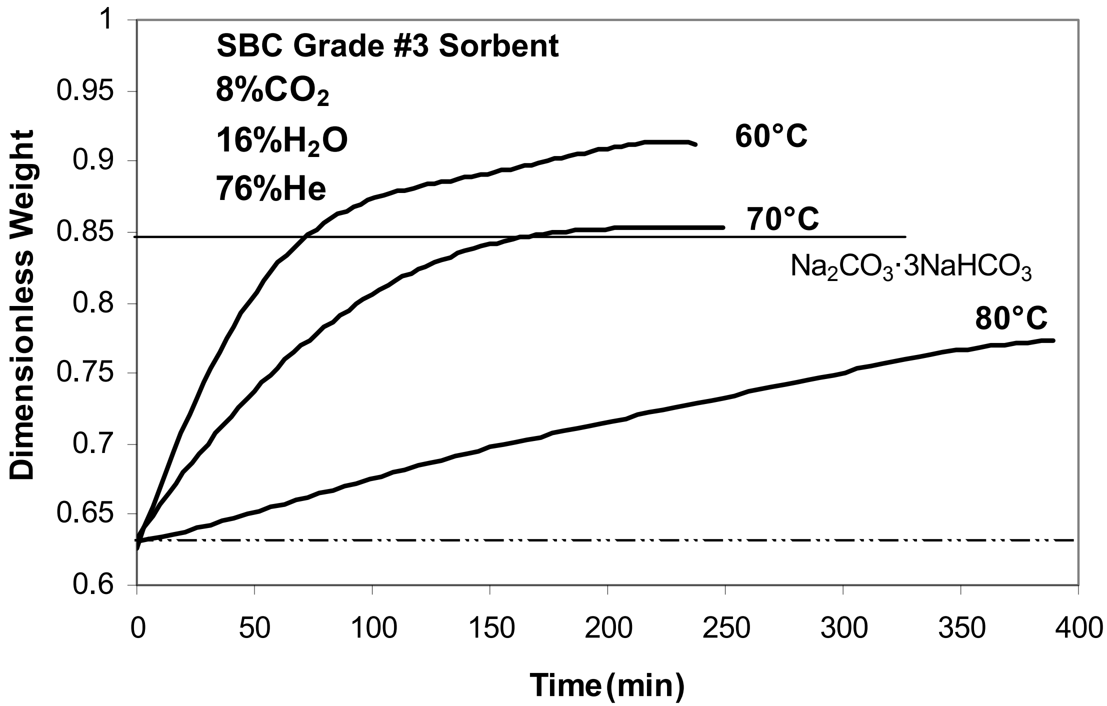
- Physical
  - Particle Size Distribution (RTI)
  - Surface Area (RTI & C&D)
  - Attrition Resistance (RTI)
  - Pore Size Distribution (RTI)
  - Bulk Density (RTI)
  - X-ray Diffraction (C&D)
  - Scanning Electron Microscopy (C&D)
  - Fluidization Characteristics (RTI)
- Chemical
  - Thermogravimetry (RTI & LSU)
  - Fixed Bed Testing (LSU)
  - Fluidized Bed Testing (RTI)

Sodium Carbonate Chemistry

Reaction	Δ H Kcal/gmol CO_2
2/3 $Na_2CO_3 \cdot 3NaHCO_3 \leftrightarrows$ 5/3 $Na_2CO_3 + CO_2 + H_2O$	32.8
5 $NaHCO_3 \leftrightarrows Na_2CO_3 \cdot 3NaHCO_3 + CO_2 + H_2O$	32.1
$2NaHCO_3 \leftrightarrows Na_2CO_3 + CO_2 + H_2O$	30.8

- CO_2 removal is exothermic
- Sorbent regeneration is endothermic

Fundamental Kinetic and Thermodynamic Studies

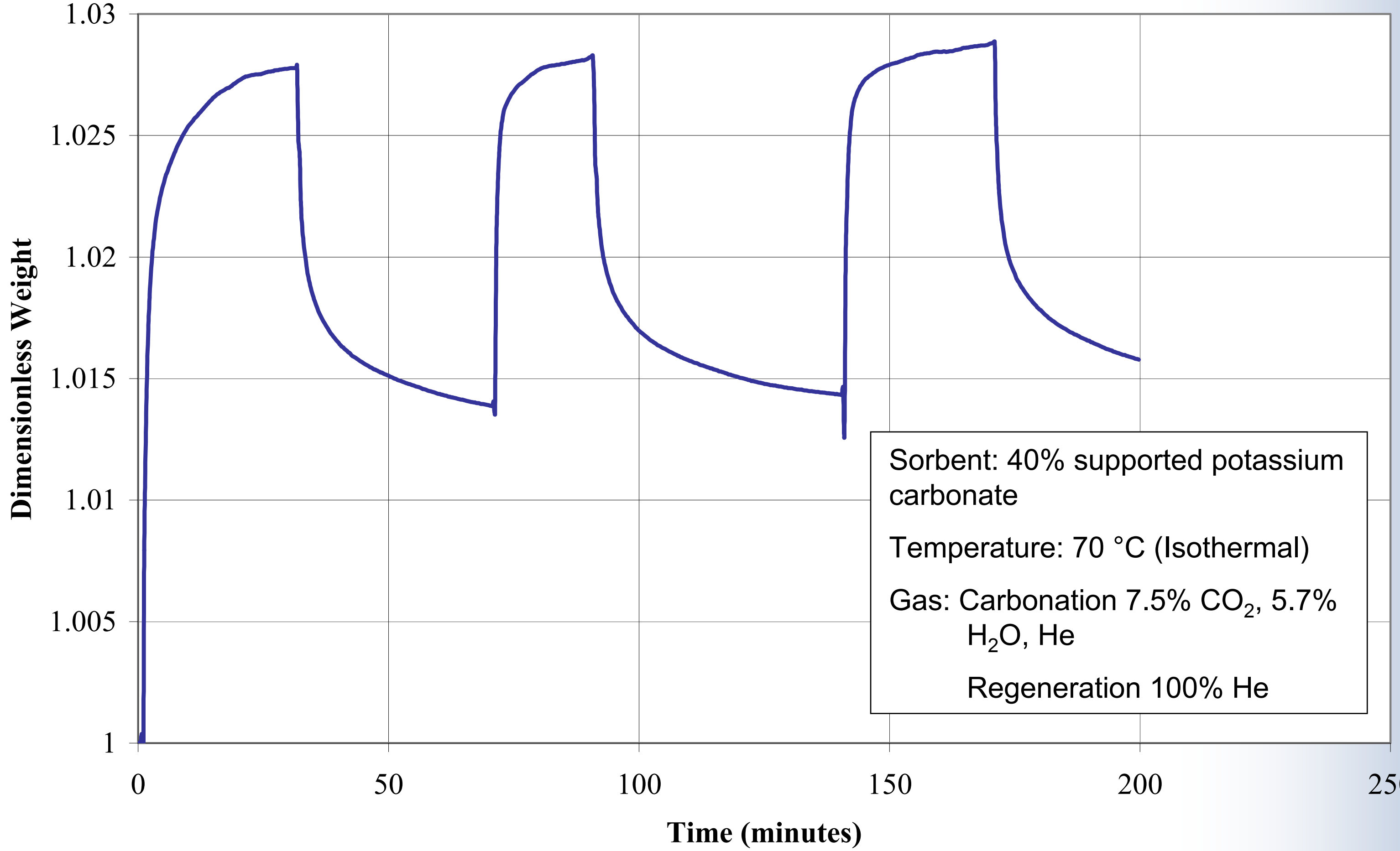


- First order reaction kinetics
  - CO_2
  - H_2O
- Temperature sensitive kinetics
  - $NaHCO_3$ product at 60 °C
  - Intermediate product (WS) at 70 °C
  - Higher temperatures decrease CO_2 removal
- Potential temperature control strategies
  - Cold diluents → solids
  - Liquid H_2O addition (ΔH_{VAP} = 10 Kcal/gmol)

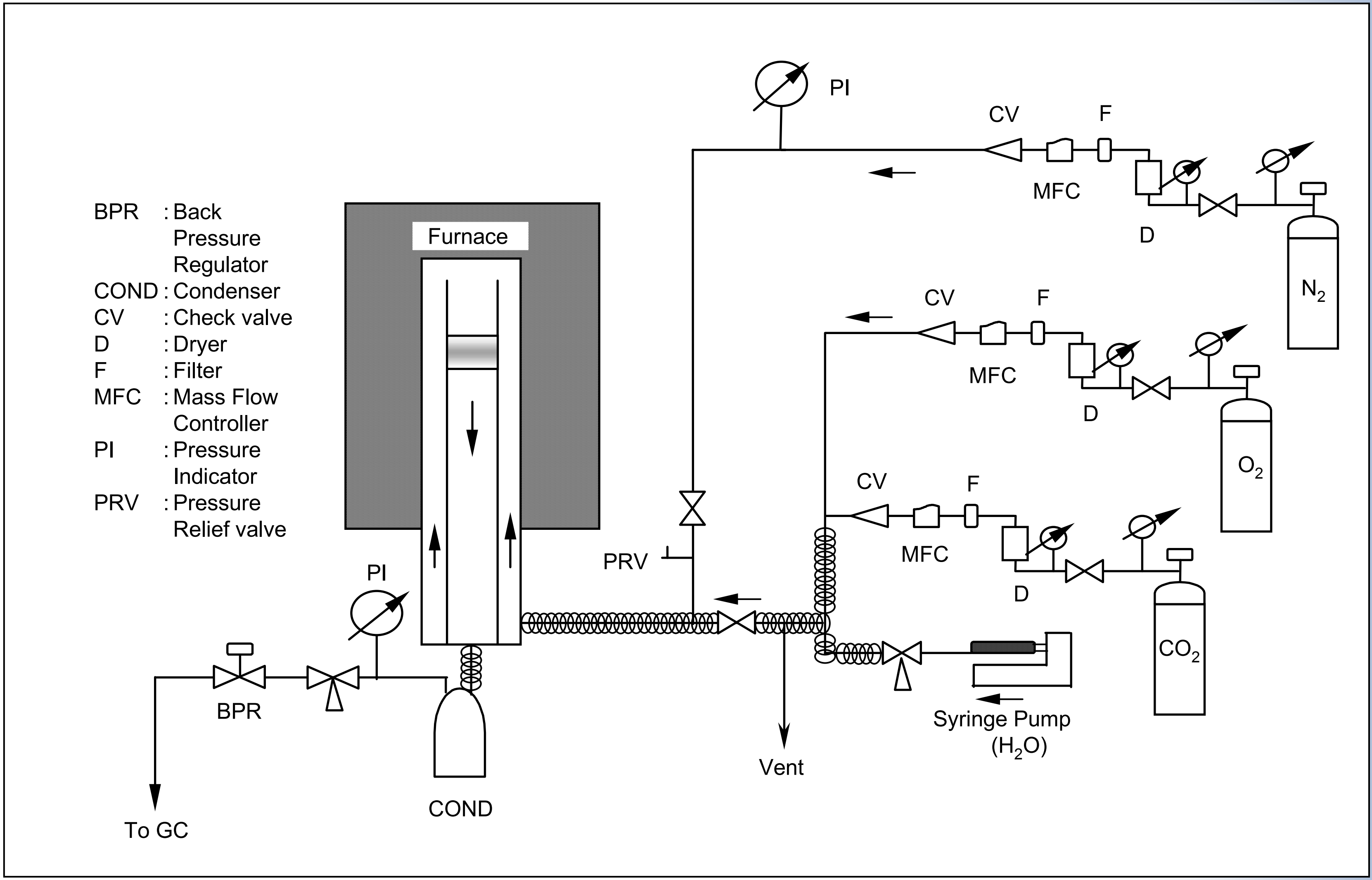
Sorbent Operating Temperature Ranges

- Sodium Carbonate
  - Carbonation: 60 – 80 °C
  - Regeneration (decarbonation; calcination): > 120 °C
- Potassium Carbonate
  - Carbonation: up to 120 °C
  - Regeneration (decarbonation; calcination): > 140 °C

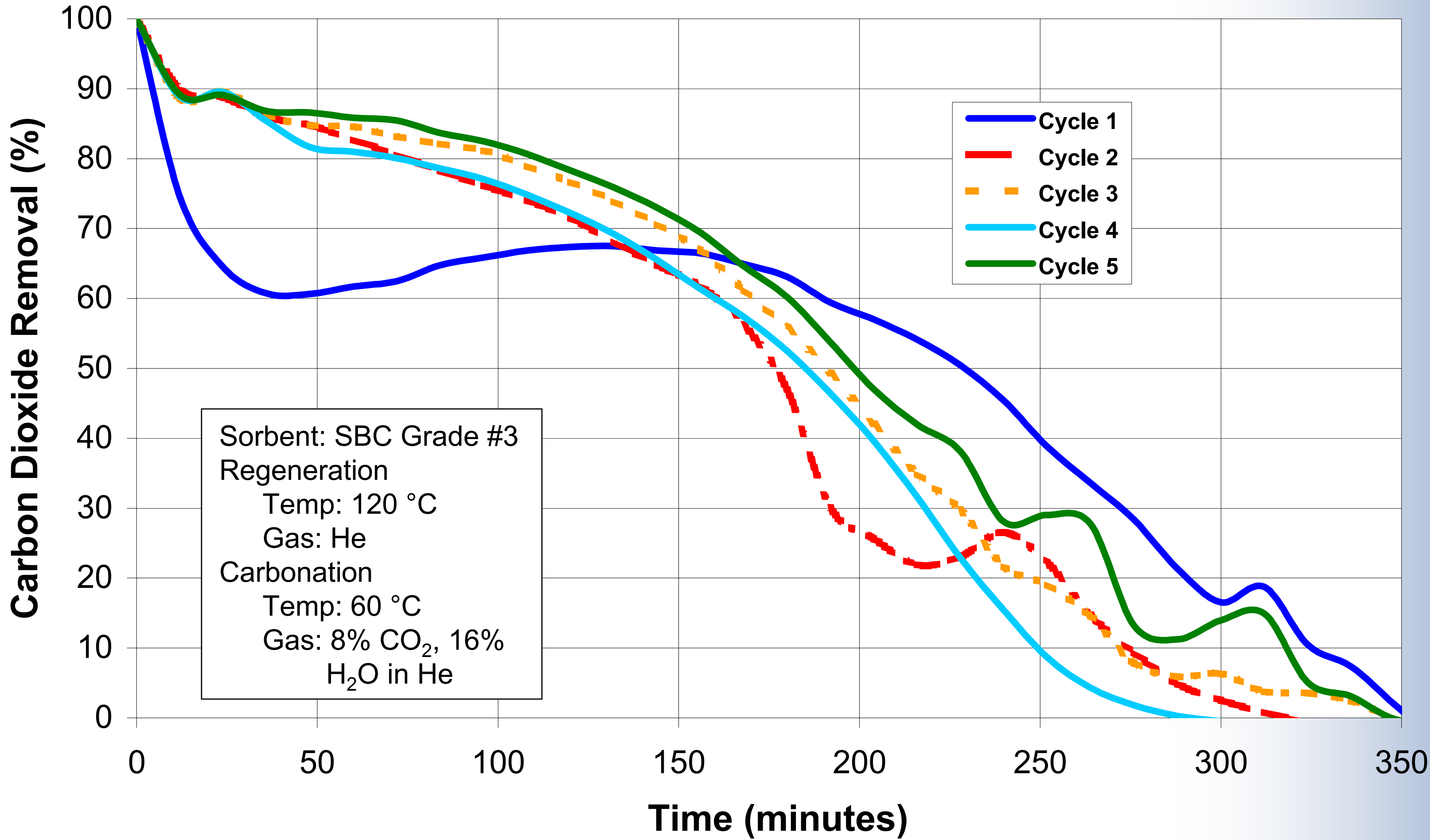
TGA Cyclic Reactivity Testing



Fixed-Bed Reactor System at LSU



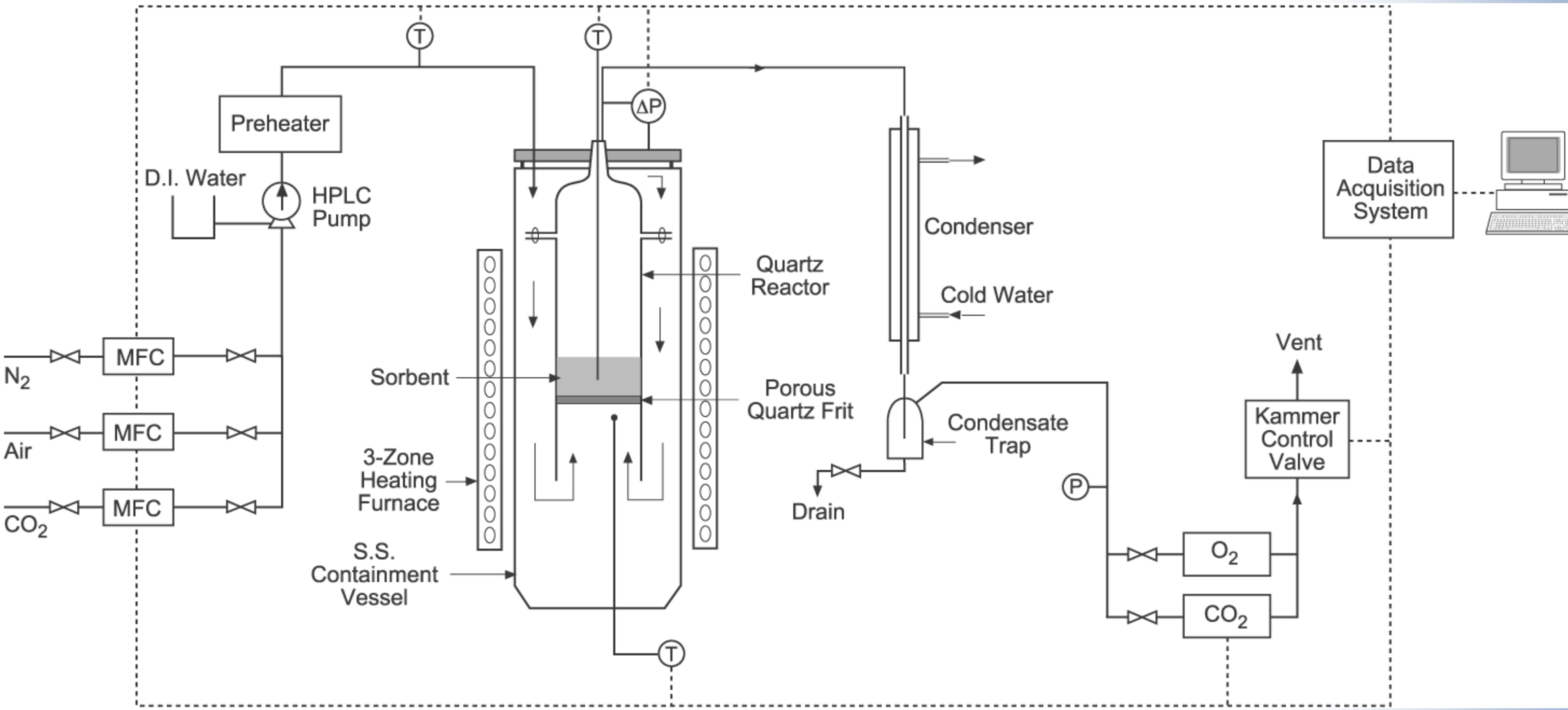
Fixed-Bed Testing of SBC



SBC Sorbent Interaction with HCl and SO_2

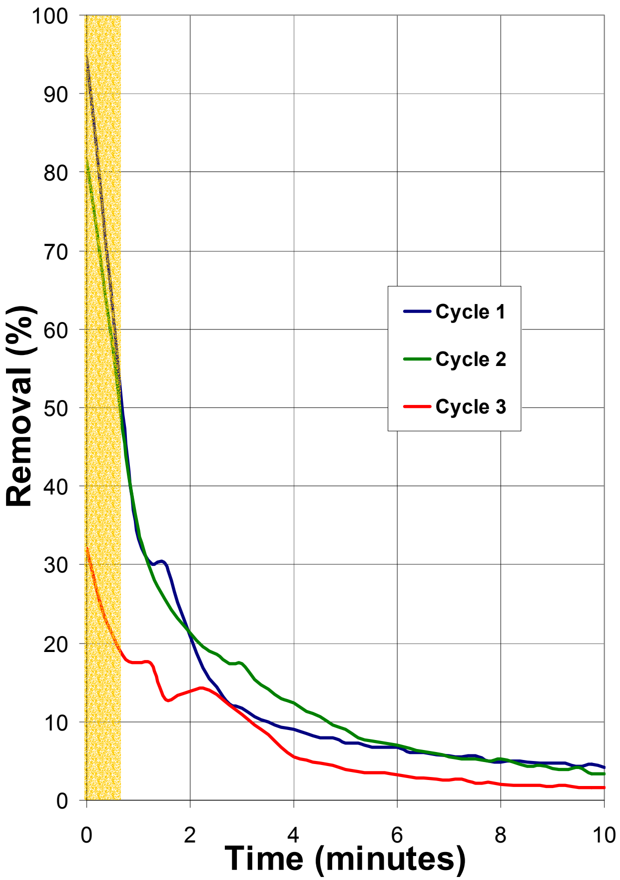
- Hydrogen Chloride
  - 1-inch Fluidized-bed testing
  - 100 ppm HCl in simulated flue gas
  - >98% removal with 1.2 sec superficial residence time
- Sulfur Dioxide
  - TGA tests and 1-inch fluidized-bed testing
  - 1000 ppm SO_2 in simulated flue gas
  - >95% removal
  - Irreversible at temperatures ≤ 200 °C

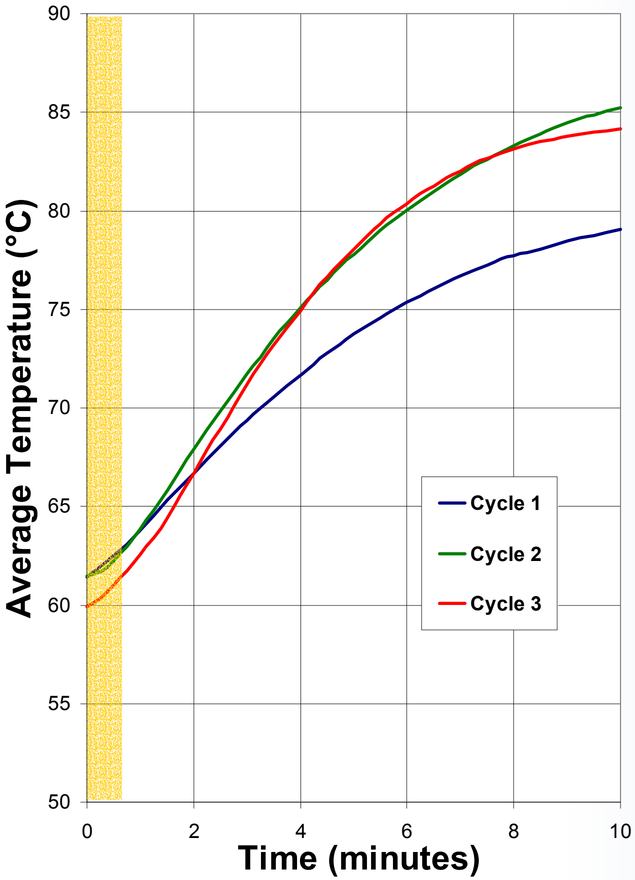
RTI's Bench-Scale Fluid-Bed Test Unit



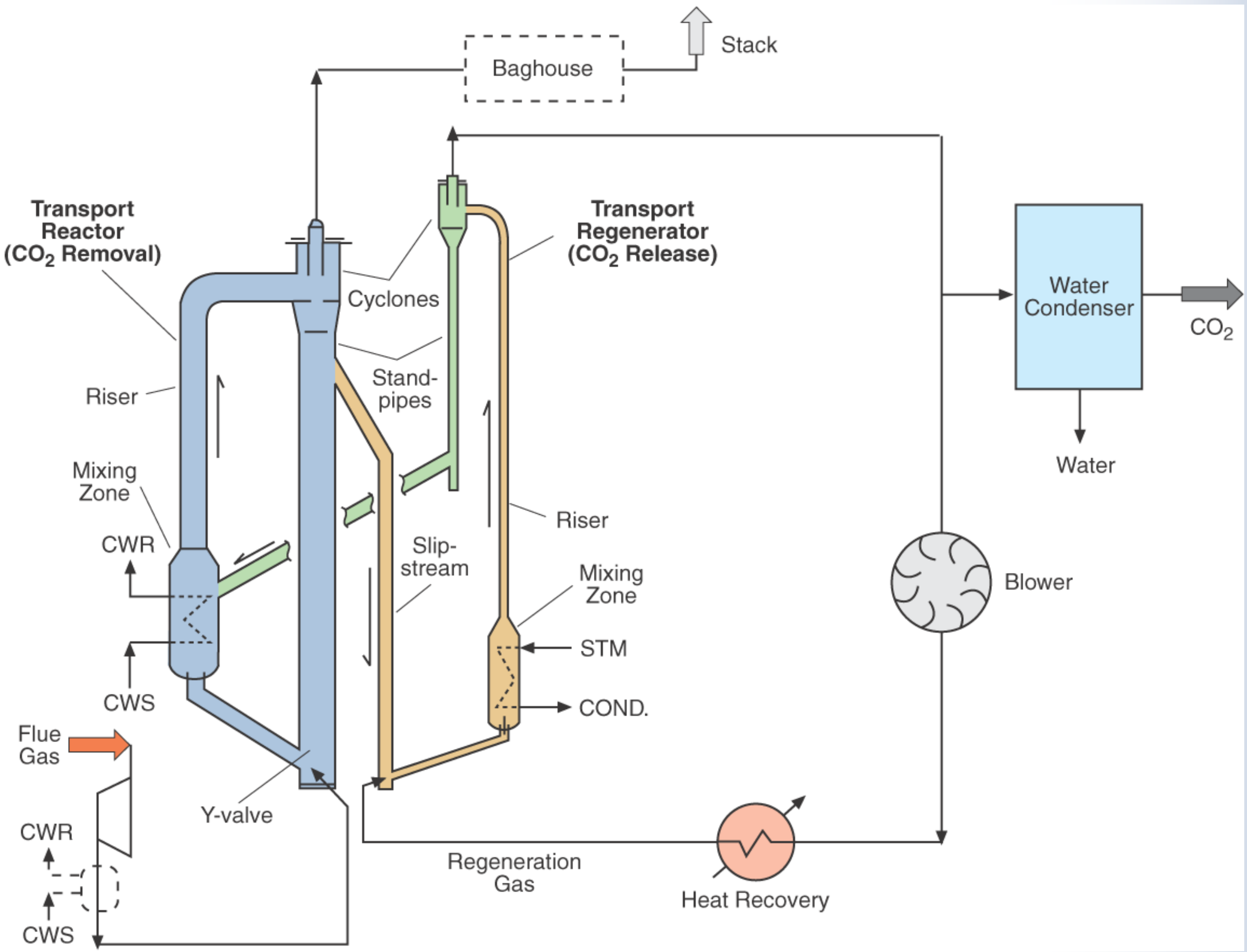
Fluid-Bed Testing of 40% Supported Sodium Carbonate

Carbonation in 7% Carbon Dioxide, 6% Water Vapor





Conceptual Transport Reactor System



Transport Reactor Approach

- Advantages
  – Low pressure drop (<1 psi [< 30 in. W.C.])
  – Reliable and effective solid sorbent movement
  – Superior temperature control
- Sorbent design challenges
  – High sorbent reactivity required
    • Short residence times (2-6 seconds)
  – Highly attrition-resistant sorbent required
    • High sorbent flux rate

Engineering Design Challenges

- Heat integration
  - Capturing low-grade, low-value heat in the steam cycle for sorbent regeneration
  - Minimizing parasitic power consumption
  - Heat transfer:
    - Removal of carbonation heat of reaction
    - Addition of regeneration energy
- Low pressure drop of flue gas stream
  - Minimizing additional power requirements of the I.D. fan
- Sorbent Transfer
  - Efficiently move sorbent between carbonation reactor and regenerator

Heat Integration Analysis

- Goal: Minimize process energy requirements
- Target: Regeneration
  - Largest energy requirement
  - Low-level heat (120-140 °C)
- Solutions
  - Steam usage
  - Low-level heat sources
    - Recover flue gas heat
    - Extract heat from cooling water
    - Alternative air preheating schemes

Comparison of Coal Fired Power Plants With and Without CO_2 Removal

Case	Heat Requirement for CO_2 Sorbent Regeneration, Btu/lbmol CO_2	Gross Plant Power kWe	Auxiliary Power Requirement kWe	Net Plant Power kWe	Plant Efficiency (HHV) %
EPRI Base Case 7C Coal Fired Steam Plant; no CO_2 Removal	Not Applicable	491,108	29,050	462,058	40.5
EPRI Case 7A MEA CO_2 Removal	71,140[E]	402,254	72,730	329,524	28.9
EPRI Case 7A Re-calc'd	103,400[A]	362,178	72,730	289,448	25.4
Comparison Case Na_2CO_3-based Dry CO_2 Removal	60,000	416,144	72,730	343,414	30.1

90% CO_2 Removal for Applicable Cases
For all cases: Heat input = 1,140,155 kW_{heat} (HHV)
[E]EPRI, Evaluation of Innovative Fossil Fuel Power Plants with CO_2 Removal, 2000
[A]Alstom Power, Engineering Feasibility and Economics of CO_2 Capture on an Existing Coal-Fired Power Plant, 2001

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Summary of Research Findings

- The sodium and potassium carbonate sorbents react readily to remove CO_2
- The materials can be cycled repeatedly without appreciable loss of activity
- The carbonate/carbon dioxide reaction may be limited by considerations of heat removal from the sorbent particle
- The high initial rates of reaction may be suitable for short residence time transport reactor systems
- Regeneration of sorbent can be carried out in an essentially pure carbon dioxide stream
- Supported materials provide suitable activity and attrition resistance

Technology Development Plan

Thermodynamic Analysis & Lab Testing

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Bench-scale Testing

Evaluate concept

Kinetic studies
Material screening
Sorbent development
Process modeling
Preliminary economics

Pilot-Plant Testing

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Slip Stream Testing

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Demonstration Testing

Scale-up of sorbent production
Sorbent evaluation
- Reactivity
- Capacity
- Attrition
- Stability

- Energy analysis
  - Heat requirements
  - Temperature constraints
- Economic evaluation

COMMERCIAL IMPLEMENTATION

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